

REMARKS

Claims 1-6 were canceled by preliminary amendment. Claims 7-28 are pending in the application; claims 25-28 have been added.

The examiner has objected to the specification because of an informality on page 6, line 13. Applicants have corrected this apparent typographical error in this communication. Support for the correction may be found in on page 7, line 30 of WO publication 01/07477.

1. Response to rejection under 35 USC §112

The examiner has rejected claims 11-19 under 35 USC §112, second paragraph, as indefinite for failing to particularly point out and distinctly claim the subject matter which the applicant regards as the invention. Under MPEP §2173.02, the examiner must consider the claim as a whole to determine whether the claim apprises one of ordinary skill in the art of its scope, and therefore serves the notice function required by 35 USC §112, second paragraph. Solomon v. Kimberly-Clark Corp., 216 F.3d 1372, 1379, 55 USPQ2d 1279, 1283 (Fed. Cir. 2000).

In this instance one of ordinary skill in the art would be apprised of the metes and bounds of the instant claims. In support of this proposition applicants' have herewith provided a copy of Ullmann's Encyclopedia of Industrial Chemistry, Volume A28 pp. 428-429. This resource provides a discussion of zeolite nomenclature. Furthermore, a database of zeolite structure may be found at <http://www.iza-structure.org/databases/> and then selecting "Atlas of Zeolite Structures." Alternatively,

this site may be reached directly at http://www.zeolites.ethyz.ch/zeolites/FMPro?-db=Atlas_main.fp5&-lay=web%20layout&-format=StdAtlas.htm&-view. An additional resource is the Atlas of Zeolite Structure Types, W.M. Meier, D.H. Olson, Ch.Baerlocher; Elsevier, 5th edition, Amsterdam 2001. (See also page 4 of the specification citing an earlier edition of this resource). The Federal Circuit explained in *S3 Inc. v. nVIDIA Corp.*, 259 1364, 59 USPQ2d 1745 (Fed. Cir. 2001),

The purpose of claims is not to explain the technology or how it works, but to state the legal boundaries of the patent grant. A claim is not 'indefinite' simply because it is hard to understand when viewed without benefit of the specification. *Cf. Autogiro Co. Of America v. United States*, 384 F.2d 391, 397, 155 USPQ 697, 701 (Ct. Cl. 1967).

In the instant specification, applicants have specifically referred to a reference of zeolite structure types and have described the types that would be appropriate for this invention. (Page 5, lines 5-27). Thus, one of ordinary skill in the art would recognize the structure types of zeolites in claims 11-14 and would understand the metes and bounds of the claims upon reading the specification.

2. Response to rejection under 35 USC §102

The examiner has rejected claims 7-19, under 35 USC §102(b) as anticipated in view of Felthouse (US 4,582, 650). The examiner argues that Felthouse discloses a process for the oxidation of N-phosphonomethyliminodiacetic acid to give N-phosphonomethylglycine utilizing a catalyst of aluminosilicates which contains a noble metal complex. However, applicants assert that Felthouse does not anticipate each and every element of the claims of the instant invention.

"To anticipate, every element and limitation of the claimed invention must be found in a single prior art reference, arranged as in the claim." Brown v. 3M, 265 F.3d 1349, 60 USPQ2d 1375 (Fed. Cir. 2001). In this instance there are significant differences in the prior art and that of the instant invention.

Felthouse is directed to catalyzing the oxidation of formaldehyde or formic acid in the presence of N-phosphonomethylglycines (NPMG). Column 1, lines 48-50.

Specifically, a noble metal catalyst is located within the pores of a zeolite or other microcrystalline support in order to avoid contact by the NPMG and other impurities.

Column 1, lines 50-55. Furthermore, Felthouse describes

The invention is particularly concerned with effecting such oxidation of formaldehyde during the production of N-phosphonomethylglycine by carbon-catalyzed oxidative cleavage of N-phosphonomethyliminodiacetic acid, as formaldehyde is a by-product in the cleavage reaction and may further react with the desired N-phosphonomethylglycine product.

Column 1, lines 55-61.

Thus, Felthouse discloses the oxidation of formaldehyde in the context of the reduction of a by-product caused by carbon-catalyzed oxidative cleavage of N-phosphonomethyliminodiacetic acid. According to the disclosure of Felthouse, the object of the oxidation is formaldehyde or formic acid. (Felthouse column 1, lines 55-61). In contrast, in the instant process PMIDE or its salt is brought into contact with at least an oxygen containing oxidant in the presence of a heterogenous catalyst comprising at least one silicate. Furthermore, according to Felthouse, the oxidation of the formaldehyde or formic acid is required as a result of the carbon catalyst used to

catalyze PMIDE to NPMG. The invention of the instant application does not utilize a carbon catalyst. Therefore, in light of the disclosure of Felthouse, one of ordinary skill in the art would interpret the reference as disclosing the oxidation of formaldehyde, a by-product of carbon-catalyzed oxidative cleavage of N-phosphonomethyliminodiacetic acid.

3. Response to rejection under 35 USC §103

The examiner has rejected claims 7-11 and 20-24 under 35 USC § 103(b) as unpatentable over Felthouse in view of applicant's admission. Specifically, the examiner argues that it would be obvious to use the process of Felthouse to prepare N-phosphonomethylglycine by oxidation of N-phosphonomethyliminodiacetic acid in the presence of a heterogeneous catalyst wherein the catalyst may be regenerated. The examiner argues that in addition to the discussion of Felthouse above, applicant's admission that catalyst regeneration is well known in the art and is therefore obvious. However, as discussed above Felthouse does not anticipate nor does it render the instant invention obvious.

As discussed below with regard to the specific references the examiner has failed to establish a *prima facie* case of obviousness with respect to the instant invention. Three requirements must be fulfilled in order for a *prima facie* case of

obviousness to be satisfied. First, there must be some suggestion or motivation in the references themselves or available to one of ordinary skill in the art to modify the reference or to combine reference teachings.¹ Second, there must be a reasonable expectation of success. Third, the prior art references combined must teach or suggest all the claim limitations. MPEP §2143. Both the suggestion to carry out the claimed process and the reasonable expectation of success must be found in the prior art and not based on the applicant's disclosure. *In re Vaeck*, 947 F.2d 488, 20 USPQ2d 1438, 1442 (Fed. Cir. 1991). Additionally, the level of ordinary skill in the art cannot be relied upon to provide the suggestion to combine references. *Al-Site Corp. v. VSI Int'l Inc.* 174 F.3d 1308, 50 USPQ2d 1161, 1171 (Fed. Cir. 1999). With respect to the instant application the examiner has failed to meet this burden.

One of ordinary skill in the art would not be motivated to produce the instant invention in view of Felthouse. The reference discloses the catalyzed oxidation of formaldehyde or formic acid in the presence of NPMG. In essence, this disclosure is directed at eliminating a by product of the main reaction wherein the main reaction is catalyzed using an activated carbon catalyst. (Felthouse column 1, lines 5-11 and 20-25). The instant process does not utilize an activated carbon catalyst. Applicants respectfully assert that the examiner has not identified the requisite motivation provided by Felthouse that would motivate one of ordinary skill in the art to adjust a two-stage

¹There are three possible sources for motivation to combine references: the nature of the problem to be solved, the teachings of the prior art, and the knowledge of persons of ordinary skill in the art. *In re Rouffet*, 149 F.3d 1350, 1357, 47 USPQ2d 1453, 1457-1458 (Fed. Cir. 1998).

process utilizing co-catalysts to a process utilizing one catalyst wherein the reference does not disclose this single catalyst as catalyzing the direct production of NPMG and PMIDE.

Furthermore, a *prima facie* case of obviousness has not been established as the examiner has failed to show that applicants' had a reasonable expectation of success. Both the Felthouse process and the process of the instant invention involve catalysis. The CCPA in *In re Doumani*, noted that catalytic effects are not ordinarily predictable with certainty. *In re Doumani*, 281 F.2d 215, 126 USPQ 408 (CCPA 1960). Felthouse does not disclose that it is possible to utilize one catalyst for the direct production of NPMG from phosphonomethyliminodiacetic acid. Accordingly, applicants respectfully assert that the examiner has failed to establish a *prima facie* case of obviousness as an expectation of success has not been sufficiently supported.

Finally, the examiner has failed to establish a *prima facie* case of obviousness as the references cited do not teach all of the claimed limitations. The oxidation of the formaldehyde or formic acid is required as a result of the carbon catalyst used to catalyze PMIDE to NPMG. (Felthouse column 1, lines 55-61). As the instant invention does not utilize carbon catalyst each limitation of the invention has not been taught by the reference. Accordingly, a *prima facie* case of obviousness cannot be supported in view of Felthouse.

In view of the foregoing amendments and remarks, applicants consider that the rejections of record have been obviated and respectfully solicit passage of the

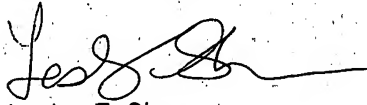
application to issue.

Please find attached a check for \$410.00 for a two month extension of time.

Please charge any shortage in fees due in connection with the filing of this paper, including Extension of Time fees to Deposit Account No. 11-0345. Please credit any excess fees to such deposit account.

Respectfully submitted,

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VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE SPECIFICATION

Please amend the paragraph found on lines 12 to 14 of page 6 as follows:

b4 Further titanium-containing zeolites which may be mentioned are those having a structure of ZSM-48 type, the ZSM-12 type, ferrierite or β β -zeolite and of mordenite.

IN THE CLAIMS

Please amend claim(s) 7 and 11-14 and add new claims 25-28 as follows:

b1 7. (currently amended) A process for preparing N-phosphonomethylglycine or a salt thereof by bringing phosphonomethyliminodiacetic acid or a salt thereof into contact with at least one oxygen-containing oxidant in the presence of a heterogeneous catalyst comprising at least one silicate, excluding carbon catalysts.

11. (currently amended) A process as claimed in claim 7, wherein the crystalline silicate or silicates is/are selected from the group consisting of zeolites of the ~~structure~~ types having framework type codes ABW, ACO, AEI, AEL, AEN, AET, AFG, AFI, AFN, AFO, AFR, AFS, AFT, AFX, AFY, AHT, ANA, APC, APD, AST, ATN, ATO, ATS, ATT, ATV, AWO, AWW, BEA, BIK, BOG, BPH, BRE, CAN, CAS, CFI, CGF, CGS, CHA, CHI, CLO, CON, CZP, DAC, DDR, DFO, DFT, DOH, DON, EAB, EDI, EMT, EPI, ERI, ESV, EUO, FAU, FER, GIS, GME, GOO, HEU, IFR, ISV, ITE, JBW, KFI, LAU, LEV, LIO, LOS, LOV, LTA, LTL, LTN, MAZ, MCM-22,

MEI, MEL, MEP, MER, MFI, MFS, MON, MOR, MSO, MTF, MTN, MTT, MTW, MWW, NAT, NES, NON, OFF, OSI, PAR, PAU, PHI, RHO, RON, RSN, RTE, RTH, RUT, SAO, SAT, SBE, SBS, SBT, SFF, SGT, SOD, STF, STI, STT, TER, THO, TON, TSC, VET, VFI, VNI, VSV, WEI, WEN, YUG, ZON and ITQ-4 and mixtures of two or more thereof.

12. (currently amended) A process as claimed in claim 8, wherein the crystalline silicate or silicates is/are selected from the group consisting of zeolites of the structure types having framework type codes ABW, ACO, AEI, AEL, AEN, AET, AFG, AFI, AFN, AFO, AFR, AFS, AFT, AFX, AFY, AHT, ANA, APC, APD, AST, ATN, ATO, ATS, ATT, ATV, AWO, AWW, BEA, BIK, BOG, BPH, BRE, CAN, CAS, CFI, CGF, CGS, CHA, CHI, CLO, CON, CZP, DAC, DDR, DFO, DFT, DOH, DON, EAB, EDI, EMT, EPI, ERI, ESV, EUO, FAU, FER, GIS, GME, GOO, HEU, IFR, ISV, ITE, JBW, KFI, LAU, LEV, LIO, LOS, LOV, LTA, LTL, LTN, MAZ, MCM-22, MEI, MEL, MEP, MER, MFI, MFS, MON, MOR, MSO, MTF, MTN, MTT, MTW, MWW, NAT, NES, NON, OFF, OSI, PAR, PAU, PHI, RHO, RON, RSN, RTE, RTH, RUT, SAO, SAT, SBE, SBS, SBT, SFF, SGT, SOD, STF, STI, STT, TER, THO, TON, TSC, VET, VFI, VNI, VSV, WEI, WEN, YUG, ZON and ITQ-4 and mixtures of two or more thereof.

13. (currently amended) A process as claimed in claim 9, wherein the crystalline silicate or silicates is/are selected from the group consisting of zeolites of the structure types having framework type codes ABW, ACO, AEI, AEL, AEN, AET, AFG, AFI,

AFN, AFO, AFR, AFS, AFT, AFX, AFY, AHT, ANA, APC, APD, AST, ATN, ATO, ATS, ATT, ATV, AWO, AWW, BEA, BIK, BOG, BPH, BRE, CAN, CAS, CFI, CGF, CGS, CHA, CHI, CLO, CON, CZP, DAC, DDR, DFO, DFT, DOH, DON, EAB, EDI, EMT, EPI, ERI, ESV, EUO, FAU, FER, GIS, GME, GOO, HEU, IFR, ISV, ITE, JBW, KFI, LAU, LEV, LIO, LOS, LOV, LTA, LTL, LTN, MAZ, MCM-22, MEI, MEL, MEP, MER, MFI, MFS, MON, MOR, MSO, MTF, MTN, MTT, MTW, MWW, NAT, NES, NON, OFF, OSI, PAR, PAU, PHI, RHO, RON, RSN, RTE, RTH, RUT, SAO, SAT, SBE, SBS, SBT, SFF, SGT, SOD, STF, STI, STT, TER, THO, TON, TSC, VET, VFI, VNI, VSV, WEI, WEN, YUG, ZON and ITQ-4 and mixtures of two or more thereof.

14. (currently amended) A process as claimed in claim 10, wherein the crystalline silicate or silicates is/are selected from the group consisting of zeolites of the structure types having framework type codes ABW, ACO, AEI, AEL, AEN, AET, AFG, AFI, AFN, AFO, AFR, AFS, AFT, AFX, AFY, AHT, ANA, APC, APD, AST, ATN, ATO, ATS, ATT, ATV, AWO, AWW, BEA, BIK, BOG, BPH, BRE, CAN, CAS, CFI, CGF, CGS, CHA, CHI, CLO, CON, CZP, DAC, DDR, DFO, DFT, DOH, DON, EAB, EDI, EMT, EPI, ERI, ESV, EUO, FAU, FER, GIS, GME, GOO, HEU, IFR, ISV, ITE, JBW, KFI, LAU, LEV, LIO, LOS, LOV, LTA, LTL, LTN, MAZ, MCM-22, MEI, MEL, MEP, MER, MFI, MFS, MON, MOR, MSO, MTF, MTN, MTT, MTW, MWW, NAT, NES, NON, OFF, OSI, PAR, PAU, PHI, RHO, RON, RSN, RTE, RTH, RUT, SAO, SAT, SBE, SBS, SBT, SFF, SGT, SOD, STF, STI,

STT, TER, THO, TON, TSC, VET, VFI, VNI, VSV, WEI, WEN, YUG, ZON and
ITQ-4 and mixtures of two or more thereof.

25. (newly added) A process for preparing N-phosphonomethylglycine or a salt thereof
by bringing phosphonomethyliminodiacetic acid or a salt thereof into contact with
at least one oxygen-containing oxidant selected from a group consisting of
hydroperoxides, gases containing molecular oxygen, oxygen-donating
compounds, nitrogen oxides and mixtures of two or more thereof;
in the presence of a heterogeneous catalyst comprising at least one silicate
selected from the group consisting of
zeolites, sheet silicates, naturally occurring or synthetically produced clay
minerals, clathrasils and mixtures of two or more thereof.

26. (newly added) A process as claimed in claim 25, wherein the zeolites are selected
from the group consisting of zeolites of the structure types having framework
type codes ABW, ACO, AEI, AEL, AEN, AET, AFG, AFI, AFN, AFO, AFR, AFS,
AFT, AFX, AFY, AHT, ANA, APC, APD, AST, ATN, ATO, ATS, ATT, ATV, AWO,
AWW, BEA, BIK, BOG, BPH, BRE, CAN, CAS, CFI, CGF, CGS, CHA, CHI,
CLO, CON, CZP, DAC, DDR, DFO, DFT, DOH, DON, EAB, EDI, EMT, EPI, ERI,
ESV, EUO, FAU, FER, GIS, GME, GOO, HEU, IFR, ISV, ITE, JBW, KFI, LAU,
LEV, LIO, LOS, LOV, LTA, LTL, LTN, MAZ, MCM-22, MEI, MEL, MEP, MER,
MFI, MFS, MON, MOR, MSO, MTF, MTN, MTT, MTW, MWW, NAT, NES, NON,
OFF, OSI, PAR, PAU, PHI, RHO, RON, RSN, RTE, RTH, RUT, SAO, SAT, SBE,

SBS, SBT, SFF, SGT, SOD, STF, STI, STT, TER, THO, TON, TSC, VET, VFI, VNI, VSV, WEI, WEN, YUG, ZON and ITQ-4 and mixtures of two or more thereof.

27. (newly added) A process as claimed in claim 25 wherein the at least one heterogeneous catalyst further comprises at least one element selected from among the elements of groups Ia, IIa, IIIa, IVa, Va, VIa, VIIa, VIIIa, Ib, IIb, IIIb, IVb, Vb, VIb, VIIb of the Periodic Table.

28. (newly added) A process as claimed in claim 25, wherein the heterogeneous catalyst is regenerated after complete or partial loss of activity and the regenerated heterogeneous catalyst is reused for preparing N-phosphonomethylglycine or a salt thereof from phosphonomethyliminodiacetic acid or a salt thereof.

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COMPLETE LISTING OF ALL CLAIMS IN THE APPLICATION

7. (currently amended) A process for preparing N-phosphonomethylglycine or a salt thereof by bringing phosphonomethyliminodiacetic acid or a salt thereof into contact with at least one oxygen-containing oxidant in the presence of a heterogeneous catalyst comprising at least one silicate, excluding carbon catalysts.
8. (previously added) A process as claimed in claim 7, wherein the oxygen-containing oxidant or oxidants is/are selected from the group consisting of hydroperoxides, gases containing molecular oxygen, oxygen-donating compounds, nitrogen oxides and mixtures of two or more thereof.
9. (previously added) A process as claimed in claim 7, wherein the silicate or silicates is/are selected from the group consisting of zeolites, sheet silicates, naturally occurring or synthetically produced clay minerals, clathrasils and mixtures of two or more thereof.
10. (previously added) A process as claimed in claim 8, wherein the silicate or silicates is/are selected from the group consisting of zeolites, sheet silicates, naturally occurring or synthetically produced clay minerals, clathrasils and mixtures of two or more thereof.
11. (currently amended) A process as claimed in claim 7, wherein the crystalline silicate or silicates is/are selected from the group consisting of zeolite-structure types having framework type codes ABW, ACO, AEI, AEL, AEN, AET, AFG, AFI, AFN,

AFO, AFR, AFS, AFT, AFX, AFY, AHT, ANA, APC, APD, AST, ATN, ATO, ATS, ATT, ATV, AWO, AWW, BEA, BIK, BOG, BPH, BRE, CAN, CAS, CFI, CGF, CGS, CHA, CHI, CLO, CON, CZP, DAC, DDR, DFO, DFT, DOH, DON, EAB, EDI, EMT, EPI, ERI, ESV, EUO, FAU, FER, GIS, GME, GOO, HEU, IFR, ISV, ITE, JBW, KFI, LAU, LEV, LIO, LOS, LOV, LTA, LTL, LTN, MAZ, MCM-22, MEI, MEL, MEP, MER, MFI, MFS, MON, MOR, MSO, MTF, MTN, MTT, MTW, MWW, NAT, NES, NON, OFF, OSI, PAR, PAU, PHI, RHO, RON, RSN, RTE, RTH, RUT, SAO, SAT, SBE, SBS, SBT, SFF, SGT, SOD, STF, STI, STT, TER, THO, TON, TSC, VET, VFI, VNI, VSV, WEI, WEN, YUG, ZON and ITQ-4 and mixtures of two or more thereof.

12. (currently amended) A process as claimed in claim 8, wherein the crystalline silicate or silicates is/are selected from the group consisting of zeolite-structure types having framework type codes ABW, ACO, AEI, AEL, AEN, AET, AFG, AFI, AFN, AFO, AFR, AFS, AFT, AFX, AFY, AHT, ANA, APC, APD, AST, ATN, ATO, ATS, ATT, ATV, AWO, AWW, BEA, BIK, BOG, BPH, BRE, CAN, CAS, CFI, CGF, CGS, CHA, CHI, CLO, CON, CZP, DAC, DDR, DFO, DFT, DOH, DON, EAB, EDI, EMT, EPI, ERI, ESV, EUO, FAU, FER, GIS, GME, GOO, HEU, IFR, ISV, ITE, JBW, KFI, LAU, LEV, LIO, LOS, LOV, LTA, LTL, LTN, MAZ, MCM-22, MEI, MEL, MEP, MER, MFI, MFS, MON, MOR, MSO, MTF, MTN, MTT, MTW, MWW, NAT, NES, NON, OFF, OSI, PAR, PAU, PHI, RHO, RON, RSN, RTE, RTH, RUT, SAO, SAT, SBE, SBS, SBT, SFF, SGT, SOD, STF, STI, STT, TER,

THO, TON, TSC, VET, VFI, VNI, VSV, WEI, WEN, YUG, ZON and ITQ-4 and mixtures of two or more thereof.

13. (currently amended) A process as claimed in claim 9, wherein the crystalline silicate or silicates is/are selected from the group consisting of zeolite-structure types having framework type codes ABW, ACO, AEI, AEL, AEN, AET, AFG, AFI, AFN, AFO, AFR, AFS, AFT, AFX, AFY, AHT, ANA, APC, APD, AST, ATN, ATO, ATS, ATT, ATV, AWO, AWW, BEA, BIK, BOG, BPH, BRE, CAN, CAS, CFI, CGF, CGS, CHA, CHI, CLO, CON, CZP, DAC, DDR, DFO, DFT, DOH, DON, EAB, EDI, EMT, EPI, ERI, ESV, EUO, FAU, FER, GIS, GME, GOO, HEU, IFR, ISV, ITE, JBW, KFI, LAU, LEV, LIO, LOS, LOV, LTA, LTL, LTN, MAZ, MCM-22, MEI, MEL, MEP, MER, MFI, MFS, MON, MOR, MSO, MTF, MTN, MTT, MTW, MWW, NAT, NES, NON, OFF, OSI, PAR, PAU, PHI, RHO, RON, RSN, RTE, RTH, RUT, SAO, SAT, SBE, SBS, SBT, SFF, SGT, SOD, STF, STI, STT, TER, THO, TON, TSC, VET, VFI, VNI, VSV, WEI, WEN, YUG, ZON and ITQ-4 and mixtures of two or more thereof.

14. (currently amended) A process as claimed in claim 10, wherein the crystalline silicate or silicates is/are selected from the group consisting of zeolite-structure types having framework type codes ABW, ACO, AEI, AEL, AEN, AET, AFG, AFI, AFN, AFO, AFR, AFS, AFT, AFX, AFY, AHT, ANA, APC, APD, AST, ATN, ATO, ATS, ATT, ATV, AWO, AWW, BEA, BIK, BOG, BPH, BRE, CAN, CAS, CFI, CGF, CGS, CHA, CHI, CLO, CON, CZP, DAC, DDR, DFO, DFT, DOH, DON,

EAB, EDI, EMT, EPI, ERI, ESV, EUO, FAU, FER, GIS, GME, GOO, HEU, IFR, ISV, ITE, JBW, KFI, LAU, LEV, LIO, LOS, LOV, LTA, LTL, LTN, MAZ, MCM-22, MEI, MEL, MEP, MER, MFI, MFS, MON, MOR, MSO, MTF, MTN, MTT, MTW, MWW, NAT, NES, NON, OFF, OSI, PAR, PAU, PHI, RHO, RON, RSN, RTE, RTH, RUT, SAO, SAT, SBE, SBS, SBT, SFF, SGT, SOD, STF, STI, STT, TER, THO, TON, TSC, VET, VFI, VNI, VSV, WEI, WEN, YUG, ZON and ITQ-4 and mixtures of two or more thereof.

15. (previously added) A process as claimed in claim 7, wherein the at least one heterogeneous catalyst further comprises at least one element selected from among the elements of groups Ia, IIa, IIIa, IVa, Va, VIa, VIIa, VIIIa, Ib, IIb, IIIb, IVb, Vb, VIb, VIIb of the Periodic Table.
16. (previously added) A process as claimed in claim 8, wherein the at least one heterogeneous catalyst further comprises at least one element selected from among the elements of groups Ia, IIa, IIIa, IVa, Va, VIa, VIIa, VIIIa, Ib, IIb, IIIb, IVb, Vb, VIb, VIIb of the Periodic Table.
17. (previously added) A process as claimed in claim 9, wherein the at least one heterogeneous catalyst further comprises at least one element selected from among the elements of groups Ia, IIa, IIIa, IVa, Va, VIa, VIIa, VIIIa, Ib, IIb, IIIb, IVb, Vb, VIb, VIIb of the Periodic Table.
18. (previously added) A process as claimed in claim 10, wherein the at least one heterogeneous catalyst further comprises at least one element selected from

among the elements of groups Ia, IIa, IIIa, IVa, Va, VIa, VIIa, VIIIa, Ib, IIb, IIIb, IVb, Vb, VIb, VIIb of the Periodic Table.

19. (previously added) A process as claimed in claim 11, wherein the at least one heterogeneous catalyst further comprises at least one element selected from among the elements of groups Ia, IIa, IIIa, IVa, Va, VIa, VIIa, VIIIa, Ib, IIb, IIIb, IVb, Vb, VIb, VIIb of the Periodic Table.
20. (previously added) A process as claimed in claim 7, wherein the heterogeneous catalyst is regenerated after complete or partial loss of activity and the regenerated heterogeneous catalyst is reused for preparing N-phosphonomethylglycine or a salt thereof from phosphonomethyliminodiacetic acid or a salt thereof.
21. (previously added) A process as claimed in claim 8, wherein the heterogeneous catalyst is regenerated after complete or partial loss of activity and the regenerated heterogeneous catalyst is reused for preparing N-phosphonomethylglycine or a salt thereof from phosphonomethyliminodiacetic acid or a salt thereof.
22. (previously added) A process as claimed in claim 9, wherein the heterogeneous catalyst is regenerated after complete or partial loss of activity and the regenerated heterogeneous catalyst is reused for preparing N-phosphonomethylglycine or a salt thereof from phosphonomethyliminodiacetic acid or a salt thereof.

23. (previously added) A process as claimed in claim 10, wherein the heterogeneous catalyst is regenerated after complete or partial loss of activity and the regenerated heterogeneous catalyst is reused for preparing N-phosphonomethylglycine or a salt thereof from phosphonomethyliminodiacetic acid or a salt thereof.
24. (previously added) A process as claimed in claim 11, wherein the heterogeneous catalyst is regenerated after complete or partial loss of activity and the regenerated heterogeneous catalyst is reused for preparing N-phosphonomethylglycine or a salt thereof from phosphonomethyliminodiacetic acid or a salt thereof.
25. (newly added) A process for preparing N-phosphonomethylglycine or a salt thereof by bringing phosphonomethyliminodiacetic acid or a salt thereof into contact with at least one oxygen-containing oxidant selected from a group consisting of hydroperoxides, gases containing molecular oxygen, oxygen-donating compounds, nitrogen oxides and mixtures of two or more thereof; in the presence of a heterogeneous catalyst comprising at least one silicate selected from the group consisting of zeolites, sheet silicates, naturally occurring or synthetically produced clay minerals, clathrasils and mixtures of two or more thereof.
26. (newly added) A process as claimed in claim 25, wherein the zeolites are selected from the group consisting of zeolites of the structure types having framework

type codes ABW, ACO, AEI, AEL, AEN, AET, AFG, AFI, AFN, AFO, AFR, AFS, AFT, AFX, AFY, AHT, ANA, APC, APD, AST, ATN, ATO, ATS, ATT, ATV, AWO, AWW, BEA, BIK, BOG, BPH, BRE, CAN, CAS, CFI, CGF, CGS, CHA, CHI, CLO, CON, CZP, DAC, DDR, DFO, DFT, DOH, DON, EAB, EDI, EMT, EPI, ERI, ESV, EUO, FAU, FER, GIS, GME, GOO, HEU, IFR, ISV, ITE, JBW, KFI, LAU, LEV, LIO, LOS, LOV, LTA, LTL, LTN, MAZ, MCM-22, MEI, MEL, MEP, MER, MFI, MFS, MON, MOR, MSO, MTF, MTN, MTT, MTW, MWW, NAT, NES, NON, OFF, OSI, PAR, PAU, PHI, RHO, RON, RSN, RTE, RTH, RUT, SAO, SAT, SBE, SBS, SBT, SFF, SGT, SOD, STF, STI, STT, TER, THO, TON, TSC, VET, VFI, VNI, VSV, WEI, WEN, YUG, ZON and ITQ-4 and mixtures of two or more thereof.

27. (newly added) A process as claimed in claim 25 wherein the at least one heterogeneous catalyst further comprises at least one element selected from among the elements of groups Ia, IIa, IIIa, IVa, Va, VIa, VIIa, VIIIa, Ib, IIb, IIIb, IVb, Vb, Vab, VIIb of the Periodic Table.
28. (newly added) A process as claimed in claim 25, wherein the heterogeneous catalyst is regenerated after complete or partial loss of activity and the regenerated heterogeneous catalyst is reused for preparing N-phosphonomethylglycine or a salt thereof from phosphonomethyliminodiacetic acid or a salt thereof.

Ullmann's Encyclopedia of Industrial Chemistry

Fifth, Completely Revised Edition

Volume A 28:

Water to Zirconium and Zirconium Compounds

Editors: Barbara Elvers, Stephen Hawkins

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Zeolites

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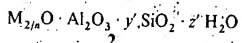
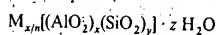
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1. Introduction

The exact definition of the term "zeolite" is still the subject of discussion [1]–[4]. A version in general use is due to BARRER [1] and BRECK [5], which states that zeolites [1318-02-1] are crystalline, hydrated aluminosilicates with a framework structure. Their three-dimensional, polyanionic networks are constructed of SiO_4 and AlO_4 tetrahedra linked through oxygen atoms. Depending on the structure type, they contain regular channels or interlinked voids whose aperture diameters are in the micropore range. These pores contain water molecules and the cations necessary to balance the negative charge of the framework. The cations, which are mobile and can be exchanged, are mainly alkali metal or alkaline earth metal ions.

Formula 1 accords with these characteristics. The expression enclosed in the square brackets

shows the composition of the anionic framework in the crystallographic unit cell. M represents the nonframework metal cation, and n is its charge. Formula 2 occurs frequently in the literature and, unlike Formula 1, simply represents the overall chemical composition.



Many synthetic zeolites also occur naturally as minerals. The observation that a certain mineral began to bubble on strong heating led CRONSTEDT in 1756 to give the name "zeolites" to such materials from the Greek *zeo*: to boil and *lithos*: stone [6], [7]. However, zeolites only became of industrial importance in the 1950s, when synthetic examples became available on an industrial scale [8]. Since then, world

consumption of synthetic zeolites in the fields of ion exchange, adsorption, and catalysis has grown continuously. The total for 1993 exceeded 10^6 t, mainly a consequence of the use of the zeolite NaA as a detergent builder. Zeolites are also important as adsorbents for separating and purifying substances and as catalysts in a number of important processes in the chemical and petrochemical industries.

In addition, ca. 300 000 t/a natural zeolites are sold worldwide. Their main uses are as ion exchangers for water treatment, in soil improvement, in animal feeds, in cat litter, and in pozzolanic cement.

The wide range of commercial applications has stimulated intensive investigation of the chemistry and physics of the zeolites [5], [9]–[19]. Current research is increasingly concerned with zeolite-like molecular sieves, which have expanded the field of zeolites enormously during the last two decades. In these new materials, the aluminum and silicon of the classical zeolites are partially or completely replaced by other elements [20], [21]. The most important groups of these molecular sieves are:

- 1) Modifications of silica ("zeosils," see → Silica, A23, p. 650)
- 2) Metallosilicates with tri- and tetravalent heteroatoms in place of aluminum as framework components, e.g., boro- and gallosilicates [22], ferrisilicates [23], and titanisilicates [24]
- 3) Alumino- and gallophosphates and compounds derived from them [25]–[27]

These compounds generally have framework structures analogous to those of the zeolites. Some new structures have been discovered, too. However, with a few exceptions, these compounds are still of rather limited economic importance, and are discussed here only briefly.

2. Nomenclature

The naming of zeolites in the literature seldom follows a scientific system. If natural analogs exist, the synthetic zeolites are often named after the minerals (e.g., faujasite, mordenite, ferrierite, and offretite). Alternatively, the names given by the discoverers are used. In the 1950s and early 1960s, new synthetic zeolites were named using the letters of the Latin and Greek alphabets. Examples include the Linde molecular sieves A, L, X, and Y of Union Car-

bide and the zeolites beta, rho, theta-1, and omega. Acronyms that indicate the industrial or university laboratory that first synthesized a given material were then increasingly used, supplemented by a consecutive numbering system. Names such as ZSM-5 (Mobil Oil), Nu-13 (ICI), LZ-210 (Union Carbide/UOP), CSZ-3 (Grace), and ECR-1 (Exxon) have gradually become generally accepted.

Zeolites with identical framework structures can therefore appear in the literature under different names. Some order has been imposed by the introduction of a classification system in the *Atlas of Zeolite Structure Types* [28], which indicates which different materials have the same structure type. The structure types themselves are denoted by the combination of three capital letters. For example, the type LTA is found in zeolite A, FAU in the faujasites X and Y, and MFI in the ZSM-5 family of zeolites.

CAS numbers are available for only a few zeolites. Moreover, as these are rarely used, they are of little benefit in literature searches.

3. Structure, Composition, and Properties of Zeolites

3.1. Framework Structure and Positions of Nonframework Cations

The third edition of the *Atlas of Zeolite Structure Types* [28], published in 1992, describes 85 different structures, including both classical zeolites and zeolite-like molecular sieves. The majority of these structures can be represented by Formulas 1 and 2, but a few have been found to occur only as alumino- or gallophosphates.

Zeolites are classified structurally as tectosilicates [3]. The basic elements of their frameworks are TO_4 tetrahedra ($\text{T} = \text{Si}, \text{Al}$) linked through oxygen atoms. The description and classification of the topology of zeolites is based on the concept of larger units known as secondary building units (SBUs) [28]. These can consist, for example, of simple rings and prisms of various sizes. By combining such basic units the known zeolite frameworks can be constructed. Alternative concepts are based on more complex chains or layer structures known as structural subunits (SSUs) [29].